GROWTH LAW OF A SPHERICAL SECOND PHASE AS GOVERNED BY SIMULTANEOUS HEAT AND MULTI-COMPONENT MASS TRANSFER LIMITATIONS-I

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Abstract-In Part I, new theoretical treatments of the growth of a stationary spherical second phase as governed by simultaneous heat and muiti-component mass transfer limitations are demonstrated. The new method is a direct extension of [l-3]. It demonstrates that the solution to these complex coupled cases can be related to the available uncoupled cases. Thus, treating the so-called "impurities" as components in the surrounding first phase, our results should include the growth of a spherical second phase as governed by simultaneous heat and mass transfer limitations in the presence of impurities as asymptotic cases.

NOMENCLATURE

$$
B_{mi}^g = (C_{wi} - C_{\infty i})/(C_{di} - C_{wi})
$$

[dimensionless];

- C_{α} mass fraction of ith solute [dimensionless];
- C_{div} **second phase ith solute mass** fraction [dimensionless] ;
- $C_{\alpha\beta}$ **ith solute mass fraction far away** from the second phase sphere [dimensionless];
- C_{n} specific heat of the surrounding first phase $\lceil \text{cal/g }^{\circ} \text{K} \rceil$;
- $C_{\text{sat1}}(T, C_2, C_3, \ldots, C_{N-1}),$ first component (main solute) saturation mass fraction $[dimensionless]$:
- $C_{\mu\nu}$ ith solute mass fraction at the interlace [dimensionless] ;
- D_{α} effective Fick's diffusion coefficient of *i*th solute $\lceil \text{cm}^2/\text{s} \rceil$:
- D/Dt , material time derivative defined in text:
- $f(t, \theta)$, function defined in equation (42), Part II;
- $\tilde{f}(t, \theta)$, function defined in equation (41), Part III;
- $G(t)$, function defined in equation (17), Part II:

 S_{\bullet}

 $T_{\rm w}$.

the center of the second phase sphere \lceil cm \rceil :

- dummy integration variable:
- time (reckoned from the com t . mencement of the growth pro- \cess $\lceil s \rceil$:
- T , $T(r, \theta, t)$, temperature $\lceil K \rceil$:
	- second phase sphere surface temperature $\lceil K \rceil$:
- T_{∞} temperature far away from the second phase sphere $\lceil K \rceil$:
- U_{∞} translational velocity of the center of the second phase sphere relative to the surrounding fluid at infinity $[cm/s]$;
- radially spherically symmetric v_{rg} convective velocity field induced by mass transfer process itself \lceil cm/s \rceil :
- radial velocity component field v_{rr} induced by the translatory motion of the second phase sphere \lceil cm/s]:
- tangential velocity component $v_{\theta t}$ field induced by the translatory motion of the second phase sphere \lceil cm/s \rceil :
- dummy integration variable : x,

 \mathcal{V} distance from the interface [cm].

Greek symbols

- x. $\equiv \lambda/(\rho C_p)$, thermal diffusivity of the surrounding first phase \lceil cm²/s]: β . growth constant dimension
	- less] :
- β _L, β calculated from the heat transfer viewpoint {dimensionless] **:**
- $\beta_{mi}^{}$ β calculated from the *i*th component mass transfer viewpoint [dimensionless]:
- ρ , density of the surrounding first phase $\lceil g/cm^3 \rceil$ *:*
- ρ_{a} density of the second phase $\lceil \frac{g}{cm^3} \rceil$:
	-

Subscripts

sphere:

 ∞ , far away from the center of the second phase sphere.

Superscripts

- 9. pertaining to multi-component second phase : \sim , pertaining to the fast moving
	- case.

INTRODUCTION

GROWTH laws of stationary, slowly moving, and fast moving spherical second phase as governed by simultaneous heat and mass transfer limitations have been obtained recently $[1-3]$. The second phase can be a bubble (gas), a droplet (liquid), or a particle (solid). In the treatments of $[1-3]$, the surrounding first phase is composed of solute and solvent, i.e. two-component environment. Now, we extend the treatment to the case when the surrounding first phase is composed of $(N - 1)$ solutes and solvent, i.e. N-component environment. Thereby, treating the so-called "impurities" as components in the surrounding first phase, our results should include the growth of a spherical second phase as governed by simultaneous heat and mass transfer limitations in the presence of impurities as asymptotic cases, as will be demonstrated later. In what follows, we will always take the first component as the main solute and Nth component as the solvent. In Part I, we treat the case when there exists no translatory motion of the center of the sphere, i.e. the spherical second phase is stationary with respect to the laboratory coordinates. In Part II, we treat the case when the spherical second phase is slowly moving. In Part III, we treat the case when the spherical second phase is fast moving. All the main results obtained in $[1-3]$ when the surrounding tirst phase is two-component environment will be recovered as the asymptotic cases. as will be demonstrated in due course.

STATEMENT OF THE PROBLEM

The problem under consideration is as follows: A spherical second phase of negligible size, i.e. $R(0) = R_0 \approx 0$, is produced in an environ-

ment, i.e. the surrounding first phase. at time $t = 0$. The second phase can be a bubble (gas). a droplet (liquid), or a particle (solid). At time $t = 0$, the entire second phase is assumed to have attained a certain proper equilibrium temperature T_{ω} , i.e. the wet bulb temperature. and remain at this temperature throughout the growth process. That is, one assumes that throughout the entire transient growth process a constant $T_{\rm w}$ exists, corresponding to a set of constant surface concentrations, $C_{\text{sat1}}(T_{w}, C_{w2})$. C_{w3} ..., C_{wN-1}) and C_{wi} (i = 2, 3, ..., $N-1$), which must be found as part of the problem solution (see Discussion). At times $t > 0$, the spherical second phase grows in the surrounding first phase due to both heat and N-component mass transfer driving forces.

The stationary spherical second phase is characterized by the following parameters: density, $\rho_{\rm d}$, latent heat of phase transition, $L(T_u)$ (< 0 for endothermic: > 0 for exothermic), and first component saturation concentration, $C_{\text{sat1}}(T, C_2, C_3, \ldots, C_{N-1})$; the surrounding first phase is characterized by the following parameters: density, ρ , specific heat, C_n , effective thermoconductivity, λ , and effective Fick's diffusion coefficients, D_i $(i = 1, 2, ..., N - 1)$. The first phase is initially at a uniform temperature T_m , and concentrations C_{mi} $(i = 1, 2, \ldots)$. $N - 1$), while the second phase is assumed to have a uniform temperature T_w and concentrations C_{di} $(i = 1, 2, ..., N - 1)$ throughout the growth process. Thus, the mass transfer process within the second phase is not considered here.

During the growth process, i.e. $t \ge 0$, the system is described by the following equations,

$$
\frac{\mathbf{D}T}{\mathbf{D}t} = \alpha \nabla^2 T, \qquad R(t) \leqslant r \leqslant \infty \qquad (1a)
$$

$$
\frac{\mathbf{D}C_i}{\mathbf{D}t} = \mathbf{D}_i \nabla^2 C_i, \qquad R(t) \leq r \leq \infty \quad (1b)
$$

with

$$
\frac{\mathbf{D}}{\mathbf{D}t} \equiv \frac{\partial}{\partial t} + \frac{R^2}{r^2} \cdot \left(1 - \frac{\rho_d}{\rho}\right) \cdot \dot{R} \cdot \frac{\partial}{\partial r}
$$

and

$$
\nabla^2 \equiv \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial}{\partial r} \right)
$$

\n
$$
T(r, 0) = T_r
$$
 (2a)

$$
C_i(r, 0) = C_{\infty i}
$$
 (2b)

$$
T(\infty, t) = T_{\infty} \tag{3a}
$$

$$
C_i(\infty, t) = C_{\infty i} \tag{3b}
$$

$$
T(R(t), t) = T_w \tag{4a}
$$

$$
C_i(R(t), t) = C_{wi}
$$
 (4b)

$$
\rho_d \vec{R} = \frac{\lambda}{-L(T_w)} \cdot \left(\frac{\partial T}{\partial r}\right)_{r=R(t)} \tag{5a}
$$

$$
\rho_d \dot{R} = \frac{D_i \rho}{C_{di} - C_{wi}} \cdot \left(\frac{\partial C_i}{\partial r}\right)_{r = R(t)} \tag{5b}
$$

$$
R(0) = R_0 \approx 0 \tag{6}
$$

where *i* runs from 1 to $N - 1$, $\alpha \equiv \lambda / (\rho C_p)$ is the thermal diffusivity of the surrounding first phase. and the first component surface concentration is assumed to be $C_{w_1} = C_{sat1}(T_w, C_{w_2},$ $C_{w,3}$ $C_{w,N-1}$). The problem is to find the a priori unknown interface temperature T_w and concentrations C_{wi} ($i = 1, 2, ..., N - 1$) and obtain the growth law of the second phase. $R(t)$.

METHOD OF SOLUTION

The key to this physically important problem is to recognize that the growth laws obtained from either heat or N-component mass transfer viewpoints must be identical. Thus. one obtains the compatibility conditions from which T_w and C_{wi} ($i = 1, 2, ..., N - 1$) are calculated (see below).

From the heat transfer viewpoint. i.e. the set of equations (1a), (2a), (3a), (4a). (5a) and (6). the temperature variable $T(r, t)$ satisfies the same boundary-value problem as in $[4-6]$. Thus, one gets $[4-6]$

$$
\frac{T-T_{\infty}}{T_{\rm w}-T_{\infty}} = \frac{\int_{\rho_1/2}^{\infty} x^{-2} \exp\left[-x^2 - 2\right.(1-\rho_d/\rho)\cdot \beta_h^3 x^{-1}\right] dx}{\int_{\beta_h}^{\infty} x^{-2} \exp\left[-x^2 - 2\right.(1-\rho_d/\rho)\cdot \beta_h^3 \cdot x^{-1}\right] dx}
$$
(7)

where

$$
R_h = 2\beta_h \cdot \sqrt{\alpha t} \tag{8}
$$

and β_k is given by with

$$
N a_h^{\theta} = - \phi \bigg(1 - \frac{\rho_d}{\rho}, \beta_h \bigg) \tag{9}
$$

with

$$
N a_h^{\theta} \equiv \frac{\rho}{\rho_d} \cdot B_h^{\theta} \equiv \frac{\rho}{\rho_d} \cdot \frac{C_p (T_{\infty} - T_{\omega})}{L(T_{\omega})} \qquad (10)
$$

and the function $\phi(1 - \rho_d/\rho, \beta)$ defined by

$$
\phi \left(1 - \frac{\rho_d}{\rho}, \beta \right) = 2\beta^3 \exp \left[\beta^2 + 2
$$

$$
\times \left(1 - \frac{\rho_d}{\rho} \right), \beta^2 \right] \cdot \int_{\beta}^{\infty} x^{-2} \exp \left[-x^2 - 2 \right]
$$

$$
\times \left(1 - \frac{\rho_d}{\rho} \right), \beta^3, x^{-1} \right] dx. \quad (11)
$$

From the ith component mass transfer viewpoint, i.e. the set of equations (lb), (2b), (3b), $(4b)$, $(5b)$ and (6) , the concentration variable $C_i(r, t)$ satisfies the same boundary-value problem as in $[4-6]$. Thus, one gets $[4-6]$

$$
N a_{mi}^{\theta} = -\phi \bigg(1 - \frac{\rho_d}{\rho}, \beta_{mi} \bigg) \tag{14}
$$

$$
N a_{mi}^{\theta} \equiv \frac{\rho}{\rho_d} \cdot B_{mi}^{\theta} \equiv \frac{\rho}{\rho_d} \cdot \frac{C_{wi} - C_{\infty i}}{C_{di} - C_{wi}} \qquad (15)
$$

and the function $\phi(1 - \rho_d/\rho, \beta)$ is given in equation (11). The uniqueness of the growth law of the second phase, i.e. $R_h(t) = R_{m1}(t) = R_{m2}(t)$ $= \ldots = R_{mN-1}(t) = R(t)$, gives the following compatibility conditions

$$
\beta_h \cdot \sqrt{\alpha} = \beta_{m1} \cdot \sqrt{D_1}
$$

= $\beta_{m2} \cdot \sqrt{D_2}$
= $-\$
= $\beta_{mN-1} \cdot \sqrt{D_{N-1}}$ (16)

where β_h and β_{mi} are given by equations (9) and (14). respectively. Owing to the $C_{\text{sat1}}(T, C_2)$ C_3, \ldots, C_{N-1}) relation. equation (16) determines a unique value of T_w and a unique set of values for C_{wi} $(i = 1, 2, ..., N - 1)$. Then, the required growth law of the second phase follows either from equations (8) - (11) or equations (13) - (15) and (11). The comprehensive plot of equations (9) or (14) was already shown in $\lceil 1 \rceil$, which is equivalent to Scriven's Fig. 1 [6].

$$
\frac{C_i - C_{\infty i}}{C_{\rm wt} - C_{\infty i}} = \frac{\int_{(r/2\sqrt{\rho_i})}^{\infty} x^{-2} \exp \{-x^2 - 2 \cdot [1 - (\rho_d/\rho)] \cdot \beta_{\rm m i}^3 x^{-1}\} \, \mathrm{d}x}{\int_{\beta_{\rm m i}}^{\infty} x^{-2} \exp \{-x^2 - 2 \cdot [1 - (\rho_d/\rho)] \cdot \beta_{\rm m i}^3 x^{-1}\} \, \mathrm{d}x}
$$
(12)

where

$$
R_{mi}(t) = 2\beta_{mi}\sqrt{(D_i t)}\tag{13}
$$

and β_{mi} is given by

In order to find the values of T_w and C_{wi} (*i =* $(1, 2, \ldots, N - 1)$, one has to use a trial-and-error method. However, in certain physically important asymptotic cases, T_w and C_{wi} $(i = 1, 2, ...)$ $\dots, N - 1$) can be readily obtained analytically (see $\lceil 1 \rceil$ for the case of $N = 2$). In particular, in case $D_1 = D_2 = ... = D_{N-1} = \alpha$, which is reasonable for a gaseous first phase. the compatibility conditions. equation (16). are exactly satisfied for any ρ_d/ρ if one merely sets B_{h}^g =

$$
B_{m1}^{g} = B_{m2}^{g} = B_{m3}^{g} = \dots = B_{mN-1}^{g}, \text{i.e.}
$$

$$
\frac{C_{p} \cdot (T_{\infty} - T_{w})^{(1)}}{L(T_{w})} = \frac{C_{w1} - C_{\infty 1}}{C_{d1} - C_{w1}}
$$

$$
= \frac{C_{w2} - C_{\infty 2}}{C_{d2} - C_{w2}}
$$

$$
= \frac{C_{wN-1} - C_{\infty N-1}}{C_{dN-1} - C_{\infty N-1}}
$$

$$
(17)
$$

where $C_{w1} = C_{sat1}(T_w, C_{w2}, C_{w3}, \ldots, C_{wN-1}).$ Owing to the $C_{\text{sat1}}(T, C_2, C_3, \ldots, C_{N-1})$ relation. equation (17) determines a unique value of T_{ω} and a unique set of values for C_{w_i} $(i = 1, 2, \ldots,$ $N - 1$). Then, the required growth law of the second phase follows either from equations (8) –(11) or (13)–(15) and (11). To fix the ideas, we will consider the following physically important asymptotic cases.

Case 1.
$$
N = 2
$$
 (18)

When $N = 2$, i.e. two-component environment, the main results obtained in [l] are recovered, as expected.

Case 2:
$$
N = 3 \tag{19}
$$

When $N = 3$, i.e. three-component environment. equation (16) degenerates into the following form

$$
\beta_{h} \cdot \sqrt{\alpha} = \beta_{m1} \cdot \sqrt{D_1} \n= \beta_{m2} \cdot \sqrt{D_2}
$$
\n(20)

where β_h and β_{mi} (i = 1, 2) are given by equations (9) and (14), respectively. Treating the first

component as the main solute. the second component as the impurity, and the third component as the solvent. this is the case when the growth of the spherical second phase is governed by simultaneous heat and mass transfer limitations in the presence of an impurity. Owing to C_{sat} (T. C_2) relation. equation (20) determines unique values for T_w , $C_{w1} = C_{sat1}(T_w, C_{w2})$, and C_{w2} . Then. the required growth law of the second phase follows either from equations $(8)+(11)$ or (13) – (15) and (11) .

DISCUSSION

Before we proceed further. we feel it is necessary to recapture the fundamental assumptions used in Part I. First of all, it is assumed that solute and heat diffusions in the surrounding first phase are adequately described by unsteady state diffusion equations with effectively constant Fick's diffusion coefficients and an effectively constant thermoconductivity. It is assumed that all the parameters characterizing second and first phases are effectively constant and there exists a local equilibrium relationship, $C_{\text{sat1}}(T_{\text{w}})$ $C_{w2}, C_{w3}, \ldots, C_{wN-1}$ at $r = R(t)$ throughout the growth process. The assumption of negligible initial size has been discussed in [I] and shown to be valid. The compatibility conditions. equation (16) are the necessary and sufficient conditions for the existence of the stated constant interface conditions solution, i.e. it guarantees the uniqueness of the growth law of the second phase, *R(t).* Thus, the basic assumption of strictly constant T_w and C_{wi} ($i = 1, 2, ..., N - 1$) is automatically justified *a posteriori* for the second phase problems of the type considered here. Physically. the necessary and sufficient compatibility conditions mean that the second phase can grow if one maintains $T(x, t) =$ T_{∞} and $C_i(\infty, t) = C_{\infty i}$ $(i = 1, 2, ..., N - 1)$ throughout the growth process.

CONCLUSIONS

In Part I. an exact treatment of the growth of a stationary spherical second phase in the presence of simultaneous heat and multi-component mass transfer limitations has been demonstrated. In general, a trial-and-error method must first be used to solve the compatibility conditions, equation (16), to obtain the *a* priori unknown second phase temperature and surface concentrations. Having thus determined T_{w_i} and $C_{w_i}(i = 1, 2, ..., N - 1)$, the growth law of the second phase is then readily obtained. Treating the so-called "impurities" as components in the surrounding first phase, our results should include the growth of a spherical second phase as governed by simultaneous heat and mass transfer limitations in the presence of impurities as asymptotic cases.

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LOI DE CROISSANCE DUNE SECONDE PHASE SPHERIQUE GOUVERNEE PAR DES CONDITIONS DE TRANSFERTS SIMULTANES DE CHALEUR ET DE MASSE-I

Résumé—On considère dans une première partie, de nouveaux traitements théoriques de la croissance d'une seconde phase sphérique et stationnaire, pilotée par des limitations de transferts simultanés de chaleur et de masse a plusieurs composants. La nouvelle mtthode est une extension directe de celle des références (1–3). Il est démontré que la solution de ces cas couplés compliqués peut être reliée à des cas non couplés connus. Ainsi, traitant les "impuretés" comme des composants dans la première phase environnante, les résultats peuvent inclure, comme des cas asymptotiques, la croissance d'une seconde phase sphérique gouvernée par des limitations de transferts simultanés de chaleur et de masse en présence des impuretés.

WACHSTUMGSGESETZ EINER KUGELFÖRMIGEN SEKUNDÄRPHASE FÜR GLEICHZEITIGEN WÄRME- UND VIEL-KOMPONENTEN-STOFFÜBERGANG---I

Zusammenfassung-Im Teil I werden neue theoretische Betrachtungen über das Wachstum einer durch gleichzeitigen Wärmeübergang und Mehrkomponentenstoffübergang begrenzten stationären kugelförmigen Sekundärphase angestellt. Die neue Methode ist eine unmittelbare Erweiterung von [1-3]. Sie zeigt, dass die Losung dieser komplex überlagerten Fälle auf vorhandene ungekoppelte Fälle übertragen werden kann. Indem man diese sogenannten "Unreinheiten" als Komponenten in der umgebenden ersten Phase behandelt, sollten unsere Beziehungen das Wachstum einer durch gleichzeitigen Wärme- und Stoffübergang begrenzten kugelförmigen Sekundärphase in Anwesenheit der Unreinheiten als asymptotische Fälle enthalten.

ЗАКОН РОСТА СФЕРИЧЕСКОЙ ВТОРОЙ ФАЗЫ ПРИ ОДНОВРЕМЕННОМ IIEPEHOCE TEILIJA M MHOTOKOMIIOHEHTHON MACCbI-I

Аннотация—В части I показана новая теоретическая трактовка роста стационарной сферической второй фазы при совместном переносе тепла и многокомпонентной массы.

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Новый метод является прямым продолжением метода, изложенного в [1-3]. Он пока-**3ывает, что для решения таких сложных задач взаимосвязанного тепло-и массообмена** можно использовать известные для простых задач решения. Таким образом, рассматривая так называемые «примеси» в качестве компонент окружающей их первой фазы, необходимо включать как асимптотический случай рост сферической второй фазы, определяемый закономерностями одновременного тепло-и массопереноса при наличии **npmeceti.**